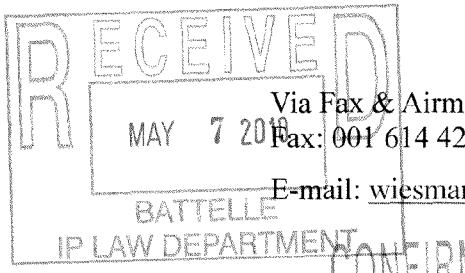


April 16, 2010

Attn.: Mr. Klaus Wiesmann  
Battelle Memorial Institute  
505 King Avenue  
Columbus, OH 43201-2693  
U.S.A.



Via Fax & Airmail  
Fax: 001 614 424 3864

E-mail: [wiesmann@battelle.org](mailto:wiesmann@battelle.org)

CONFIRMATION

**Re: National Phase in China of PCT/US2005/001402**  
In the name of Battelle Memorial Institute  
Title: Methods and Apparatus for Producing Ferrate (VI)  
CN Application No.: 200580002471.5  
Your Ref.: 22114(1)CN  
Our Ref.: 080600408

Dear Mr. Wiesmann:

In connection with the above-referenced application, the State Intellectual Property Office [SIPO] of the PRC has issued the First Office Action, which is enclosed with its English translation.

In summary, the Examiner points out that there lacks unity between independent claim 1 and each of independent claims 46, 62, 93, 96, and between independent claims 25 and 78, which is not in conformity with the provisions of Article 31(1) of Patent Law; and compared with Reference 1 (US4256551), claims 1-4, 6-7 and 12-18 do not possess novelty under Article 22(2) of Patent Law; claims 4-13 and 19-24 do not possess inventiveness under Article 22(3) of Patent Law. Please see the attached text of the First Office Action for the Examiner's detailed opinions. We also would like to provide the following further information for your reference.

Regarding the issues of novelty and inventiveness,

Pursuant to the related regulations about novelty in China, generally if the technical scheme of an invention is substantively same as that of one reference, and a person having ordinary skills in the art could determine that both technical schemes can apply to the same technical field, to solve the same technical problem and achieve the same technical effect, this invention has no novelty.

Therefore, regarding the issue of novelty in this case, we suggest comparing this invention with Reference 1 mainly in technical scheme, and stating the distinguishing technical features in technical schemes. If the technical fields, technical problems to be solved and technical effects achieved, especially technical fields, are different, the response should include such aspects too.

Furthermore, according to Chinese practice, on the basis that the novelty of the invention has been proved, it is also necessary to prove the inventiveness of the invention. As for the inventiveness issue, please see our suggestion below for your reference.

Pursuant to the related regulations about inventiveness in China, generally an Examiner works on three aspects: (1) finding a closest 1<sup>st</sup> technical scheme in the art (1<sup>st</sup> reference) to determine distinguishing features of an invention; (2) finding a 2<sup>nd</sup> technical scheme in the art (another part of 1<sup>st</sup> reference, or 2<sup>nd</sup> reference, or common knowledge in the art) to prove that 2<sup>nd</sup> technical scheme discloses related techniques and the related techniques act the same functions as the distinguishing features in the invention, which is deemed to provide a teaching from 1<sup>st</sup> technical scheme or a combination of 1<sup>st</sup> and 2<sup>nd</sup> technical schemes to apply the distinguishing features to the 1<sup>st</sup> technical scheme to solve the technical problem solved by the distinguishing features of the invention, concluding obviousness; (3) determining whether the invention provides beneficial technical effects.

Therefore, regarding the issue of inventiveness in this case, we suggest finding out (1) distinguishing features of this invention by comparing with Reference 1, (2) whether Reference 1 does disclose the similar or same techniques related to the distinguishing features, whether the disclosed techniques act the same functions as the distinguishing features in the invention, or whether the distinguishing features belong to common technical knowledge of the art, (3) the beneficial technical effects provided by the invention; and then preparing arguments on the above information.

If claims in this invention indeed do not possess novelty or inventiveness in regard to the cited Reference, we suggest making amendments to them so as to make them acquire novelty or inventiveness. If the applicant wishes, upon receiving your instructions, we can do the comparison, provide analysis and proposed amendments and prepare the response. Our attorney fee is based on the time spent in handling this matter; the billing rate of the attorney handling this matter would be USD125 per hour.

Regarding the issue of unity, to determine whether two or more inventions claimed in an application meet the requirement of unity in accordance with Article 31(1) and Rule 35 is to determine whether the substantive contents of the technical solution described in the claims belong to a single general inventive concept, that is, to determine whether these claims contain one or more of the same or corresponding particular technical features which make the claimed inventions technically interrelated, wherein said particular technical features mean those technical features that define a contribution which each of the inventions, considered as a whole, makes over the prior art. Therefore, one or more of the same or corresponding particular technical features must be present in each of independent claims in order to show unity.

Therefore, generally there are two options for solving this defect: (1) retaining the claims possessing the unity and deleting those without unity; (2) retaining the claims possessing the unity and amending the others by adding at least one particular technical feature same as or corresponding to that in the retained claims.

As for the present application, it is recommended to argue the novelty and inventiveness of claim 1 and its dependent claims without or with appropriate amendment first, and then

retain the claims possessing the unity and amend the others by adding at least one particular technical feature same as or corresponding to that in the retained claims.

Please note that in a response to the OA, the amendments made to the claims must not go beyond the scope described in the initial description and claims. In China, whether an amendment goes beyond the scope is a big issue, and the examiners will examine this issue strictly, much stricter than examiners in many other countries. **Any amendment should be found literally in the initial description and claims or determined directly and undoubtedly from the drawings and the contents literally recorded in the initial description and claims.**

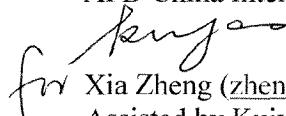
A response and possible amendments must be submitted to the Office by **July 25, 2010 (Beijing Time)**. Please give us your further instructions as soon as possible. The above official deadline can be extended once for one or two months. The cost for a one month extension is RMB940.00, including the official fee of RMB300.00 and an attorney fee of RMB640.00. The cost for a two-month extension is RMB1240.00, including the official fee of RMB600.00 and an attorney fee of RMB640.00.

We also list related rules of Chinese Patent Law below for your reference.

Should you have any further questions, please do not hesitate to contact us. We look forward to hearing from you.

Sincerely yours,

AFD China Intellectual Property

  
for Xia Zheng ([zhengxia@afdip.com](mailto:zhengxia@afdip.com))  
Assisted by Kuiyan Zhang ([info@afdip.com](mailto:info@afdip.com))

Ylh/Lc/Knx

**Enclosures:**

1. The Notification of the First Office Action (13 pages-by Airmail only);
2. English translation of the First Office Action (8 pages-by Fax & Airmail); and
3. Our invoice No. IP20101106 (1 page-by Fax & Airmail).

**Patent Law:**

**Article 22(2):** Novelty means that the invention or utility model does not belong to prior art; nor has any other party or individual filed previously with the Patent Administration Department Under the State Council an application which described the identical invention or utility model before the date of filing and was recorded in patent application documents published after said date of filing or was announced in patent documents after said date of filing.

**Article 22(3):** Inventiveness means that, as compared with the prior art, the invention possesses prominent substantive features and represents a notable progress and that the utility model possesses substantive features and represents progress.

**Article 31(1):** An application for a patent for invention or utility model shall be limited to one invention or utility model. Two or more inventions or utility models belonging to a single general inventive concept may be filed as one application.

**Article 33:** An applicant may amend his or its application for a patent, but the amendment to the application for a patent for invention or utility model may not go beyond the scope of the disclosure contained in the initial description and claims, and the amendment to the application for a patent for design may not go beyond the scope of the disclosure as shown in the initial drawings or photographs.

## Text of the First Office Action

**(For the National Phase of the PCT Application)**

Application No.: 2005800024715

The present application relates to electrochemical cells and methods of operating the same, as well as methods for making ferrate. Upon examination, [the Examiner] now provides the following examination opinions.

The present application includes the following seven groups of claims: I (1-24), II (25-45), III (46-61), IV (62-77), V (78-92), VI (93-95) and VII (96).

***I. There lacks unity between independent claim 1 and claims 46, 62, 93, 96, which is not in conformity with the provisions of Article 31(1) of Patent Law.***

The same or corresponding technical feature between claim 1 and claims 46, 62 is: an undivided electrochemical cell (an electrochemical cell) comprising a cathode, an anode and an electrolyte solution. However, the undivided cell comprising a cathode, an anode and an electrolyte solution is a common cell in the relevant field of technology and is a common technical means in the field of electrochemistry. Therefore, there is no unity between claims 1 and 46 and between claims 1 and 62.

The same or corresponding technical feature between claim 1 and claims 93, 96 is: a housing defining an undivided chamber, the housing having an electrolyte inlet, electrolyte outlets, an anode, a cathode and an electrolyte. However, it is a conventional technical means in the field of electrochemistry that a cell has a housing defining an undivided chamber, within which, an anode, a cathode and an electrolyte are contained and an electrolyte inlet and outlets are provided. Therefore, there is no unity between claims 1 and 93 and between claims 1 and 96.

The Examiner has conducted search and examination for claim 1 as well as its dependent claims. The applicant shall delete claims 46, 62, 93 and 96 which do not possess unity with respect to claim 1 and have not been searched, so as to make the present application meet the requirements of Article 31(1) of Patent Law. The applicant should note that if the amendment the applicant makes to the application documents only involves deleting claim 1 which has been searched and commented while retaining claims 46, 62, 93 and 96 as well as their dependent claims which do not possess unity with respect to claim 1 and have not been searched, such amendment is not in conformity with the provisions of Rule 51(3) of the Implementing Regulations of Patent Law. In such condition, the Examiner will, based on the application text currently examined, issue a Decision of Rejection on the ground that the present application is not in conformity with the provisions of Article 31(1) of Patent Law.

***II. Based on the current application text, the examination opinions regarding***

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 2010.2 Electronic Application: Related documents shall be submitted in the form of E-documents via the electronic patent application system. Unless otherwise provided, documents submitted in paper or other forms shall be deemed as not submitted.

*independent claim 1 as well as its dependent claims are set forth as follows:*

Claims 1-4, 6-7 and 12-18 do not possess novelty under Article 22(2) of Patent Law; and

Claims 4-13 and 19-24 do not possess inventiveness under Article 22(3) of Patent Law.

1. Claim 1 seeks protection for an undivided electrochemical cell. Reference 1 (US4256551) disclosed an undivided electrochemical cell without membrane therein, comprising a housing defining an undivided chamber, the housing having an electrolyte inlet port 7, an outlet port 8 for liquid chlorine along with brine, an outlet port 9 for sodium hydroxide solution along with brine and an outlet port 10 for hydrogen, the chamber being provided with a cathode 17 and an anode 18 and filled with an electrolyte, wherein said cathode may be nickel or mild steel mesh and said anode may be titanium mesh (both the anode and the cathode are not gas diffusion electrodes) (see the abstract; columns 1-2 and the contents from line 39 in column 4 to line 42 in column 5 of the description; and FIGS. 1, 2 and 3 of Reference 1). Thus it can be seen that Reference 1 has disclosed all the technical features of claim 1 and their technical schemes are the same; additionally, the technical scheme disclosed by Reference 1 and that sought for protection in claim 1 belong to the same technical field, can solve the same technical problem, and can produce the same technical effect. Therefore, the technical scheme sought for protection in claim 1 does not possess novelty.

2. Claim 2 depends on claim 1, and its additional technical feature further defines the electrolyte outlets; however, said additional technical feature also has been disclosed by Reference 1 (see lines 22-42 in column 5 of the description and FIGS. 1, 2 and 3 of Reference 1): the cell housing was provided with an outlet port 8 for liquid chlorine along with brine and an outlet port 9 for sodium hydroxide solution along with brine, i.e., it had two electrolyte outlets. Therefore, when the cited claim does not possess novelty, claim 2 does not possess novelty either.

3. Claim 3 depends on claim 1 and claim 4 depends on claim 3, but their additional technical features also have been disclosed by Reference 1 (see lines 1-42 in column 5 of the description and FIGS. 1, 2 and 3 of Reference 1): the cell was provided with a splitter 19 (serving as a fluid flow controller of a weir structure) in fluid communication with the electrolyte outlets, controlling the flow of the electrolyte through the outlets to control the flow of the brine electrolyte over the surfaces of the cathode and the anode. Therefore, when the cited claim does not possess novelty, claims 3 and 4 do not possess novelty either. Meanwhile, selecting the fluid flow controller from flow restrictions, valves, screens, fluid flow constrictions or bends only relates to common option in the relevant field of technology; that is to say, when the above fluid flow controllers are selected, claim 4 does not possess inventiveness.

4. Claim 5 depends on claim 1, and its additional technical feature further defines the material of the anode in the cell. However, to a person having ordinary skill in the art, using a material containing iron to make the anode is a common selection in the relevant field of

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technology, and it does not require creative work. Therefore, when the cited claim does not possess novelty, claim 5 does not possess inventiveness.

5. Claims 6 and 7 depend on claim 1, and their additional technical features further define the anode in the cell. However, said additional technical features also have been disclosed by Reference 1 (see lines 18-31 in column 2 and lines 1-9 in column 5 of the description of Reference 1): the anode in the cell may be titanium mesh (a subordinate concept of expanded metal mesh), for example, it may be a titanium mesh coating titanium oxide or ruthenium oxide (a DSA electrode). Therefore, when the cited claim does not possess novelty, claims 6 and 7 do not possess novelty either. Furthermore, it is easy for a person having ordinary skill in the art to think of selecting the anode from solid iron plate, wire mesh, woven metal cloth, wire, rod, or combinations thereof as well as iron, steel, platinum, iridium, and other oxidation resistant electrolytically conductive materials; namely, claims 6 and 7 do not possess inventiveness.

6. Claims 8 and 9 both depend on claim 1, and claims 10 and 11 both depend on claim 9. Their additional technical features further define the relative surface area of the anode to the cathode. However, a person having ordinary skill in the art can obtain, by means of regular experimental adjustment, corresponding ratio of the surface area of the anode to that of the cathode as defined in said additional technical features, and it does not require creative work. Therefore, when the cited claims do not possess novelty/inventiveness, claims 8, 9, 10 and 11 do not possess inventiveness.

7. Claims 12 and 13 depend on claim 1, and their additional technical features further define the cathode in the cell. However, said additional technical features have been disclosed by Reference 1 (see lines 1-9 in column 5 of the description of Reference 1): the cathode in the cell may be nickel or mild steel mesh. Therefore, when the cited claim does not possess novelty, claims 12 and 13 do not possess novelty either. Meanwhile, when the cathode is made of other materials or in other structures as recited in the additional technical features, claims 12 and 13 do not possess inventiveness, since they can be obtained through regular selection in the relevant field of technology.

8. Claim 14 depends on claim 1; claims 15, 16 and 17 depend on claim 14; and claim 18 depends on claim 17. Their additional technical features further define the working solution in the cell, but in Reference 1 the cell is also filled with said working solution, so the limitation to the working solution in the additional technical features cannot distinguish the cell structure for which protection is sought in the present application from the cell of Reference 1. Therefore, when the cited claim does not possess novelty, claims 14-18 do not possess novelty either.

9. Claim 19 depends on claim 1; claims 20 and 21 depend on claim 19; and claim 22 depends on claim 20. In the cell disclosed by Reference 1, a splitter is provided to vary the flow of the electrolyte over the surfaces of the cathode and the anode, for example, the flow over the surface of the anode may be controlled at 40ml/min and the flow over the surface of

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the cathode may be controlled at 30ml/min (see lines 1-42 in column 5 of the description and the Example of Reference 1). In order to provide a better control over different flow of electrolyte over the surfaces of the cathode and the anode, it is easy for a person having ordinary skill in the art to think of providing between the anode and the cathode of the cell a meshed screen; and in order to resist alkaline corrosion, it is a common selection in the art to use the plastic selected from polyolefins, fluoropolymers, or polyvinyl chloride as the material of the screen, and it does not require creative work; meanwhile, the mesh on the screen provides an open area between the cathode and the anode, the size of the mesh can be obtained by a person having ordinary skill in the art through limited times of experimental adjustment. Therefore, when the cited claim does not possess novelty/inventiveness, claims 19-22 do not possess inventiveness.

10. Claim 23 depends on claim 1, and its additional technical feature further defines the housing of the cell. Reference 1 disclosed that glass or silica can be selected to fabricate the baseplate, walls and cover of the cell housing (see lines 51-68 in column 4 of the description of Reference 1). On the basis of the above disclosure, it is easy for a person having ordinary skill in the art to think of making the housing using a material selected from metal, fiberglass, thermoplastic reinforced plastic or thermoset reinforced plastic, concrete, rubber, or combinations thereof, including constructed as a corrosion resistant liner within rigid-walled structure. Therefore, when the cited claim does not possess novelty, claim 23 does not possess inventiveness.

11. Claim 24 depends on claim 1, and its additional technical feature further defines the power supply. However, it is a common technical means for a person having ordinary skill in the art to select a variable direct current power supply operatively connected to the electrochemical cell, which does not require creative work. Therefore, when the cited claim does not possess novelty, claim 24 does not possess inventiveness.

***III. There lacks unity between independent claims 25 and 78, which is not in conformity with the provisions of Article 31(1) of Patent Law.***

The same or corresponding technical feature between claim 25 and claim 78 is: the electrochemical cell comprising a housing defining an undivided chamber, the housing having an electrolyte inlet, at least two electrolyte outlets, an anode in the chamber, and a cathode in the chamber; introducing an electrolyte through the inlet; and controlling an amount of electrolyte flowing out of the electrolyte outlets so that more electrolyte flows past the anode than the cathode. By referring to the examination opinions stated in Items II.1, II.3 and II.9, it can be known that the above technical features have been disclosed by Reference 1; namely, they are not the technical features defining a contribution over the prior art. Therefore, there is no same or corresponding particular technical feature between claims 25 and 78, and thus there lacks unity.

The applicant should make a response within the time limit specified in this notification to answer all the issues raised herein and amend the application documents when necessary;

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otherwise, the present application can hardly be granted patent right. The amendments made to the application documents must not go beyond the scope recorded in the initial description and claims as required by Article 33 of Patent Law.

The amendment documents to be submitted should include: a copy of part of the original document where amendments are involved, on which, the amendments (adding, deleting or replacement) should be marked; and re-printed substitute sheets (in duplicate) for replacing the corresponding part of the original document. The applicant should ensure consistency of contents between the above two documents.

Examiner's Name: Fangfang Peng

Examiner's Code: 199121

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**THE STATE INTELLECTUAL PROPERTY OFFICE OF THE PEOPLE'S REPUBLIC OF CHINA**

Suhua Lu, Xia Zheng AFD China Intellectual Property Suite B 1601A, 8 Xueqing Rd. Haidian District, Beijing Postal Code: 100192	Issue Date:  <b>March 10, 2010</b>
<b>Application/Patent No.: 200580002471.5</b> <b>Issue No.: 2010030600050260</b>	
<b>Applicant/Patentee:</b> Battelle Memorial Institute	
<b>Title of Invention:</b> Methods and Apparatus for Producing Ferrate (VI)	

**THE FIRST OFFICE ACTION**

(For the National Phase of the PCT Application)

1.  This application has been examined as to substance by the Office pursuant to Article 35(1) of Patent Law upon the applicant's request for substantive examination.  
 This application has been examined as to substance by the Office pursuant to Article 35(2) of Patent Law upon the Office's own decision.
2.  The applicant claims:  
the filing date January 16, 2004 in the US Patent Office as the priority date.
3.  Upon examination, the amendment submitted by the applicant on \_\_\_\_\_ is not in conformity with the provisions of Rule 51(1) of the Implementing Regulations of Patent Law and is thus not accepted.
4.  The examination is made on the Chinese text or Chinese translation of the international application originally filed.
5.  The following reference documents have been cited in this office action (their serial numbers will be referred to in the ensuing examination procedure):

Serial No.	Reference document number or title	Publication Date (or Filing date of interference patent applications)
1	US4256551	March 17, 1981

6. The conclusive opinion of the examination is as the following:

Regarding the description:

- the subject matter of the application falls into the unpatentable scope prescribed by Article 5 of Patent Law.
- the description is not in conformity with the provisions of Article 26(3) of Patent Law.
- the description is not in conformity with the provisions of Article 33 of Patent Law.
- the description is not drafted in accordance with the provisions of Rule 17 of the Implementing Regulations of Patent Law.
- \_\_\_\_\_

Regarding the claims:

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(Translated by AFD)

## THE STATE INTELLECTUAL PROPERTY OFFICE OF THE PEOPLE'S REPUBLIC OF CHINA

- claims \_\_\_\_ are not in conformity with the provisions of Article 2(2) of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Article 9(1) of Patent Law.
- claims 1-4, 6-7 and 12-18 do not possess novelty prescribed by Article 22(2) of Patent Law.
- claims 4-13 and 19-24 do not possess inventiveness prescribed by Article 22(3) of Patent Law.
- claims \_\_\_\_ do not possess practical applicability prescribed by Article 22(4) of Patent Law.
- claims \_\_\_\_ fall into the unpatentable scope prescribed by Article 25 of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Article 26(4) of Patent Law.
- claim 1 and claims (46, 62, 93, 96), claim 25 and claim 78 are not in conformity with the provisions of Article 31(1) of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Article 33 of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Rule 19 of the Implementing Regulations of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Rule 20 of the Implementing Regulations of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Rule 21 of the Implementing Regulations of Patent Law.
- claims \_\_\_\_ are not in conformity with the provisions of Rule 22 of the Implementing Regulations of Patent Law.
- \_\_\_\_\_
- The application is not in conformity with the provisions of Article 26(5) of Patent Law or Rule 26 of the Implementing Regulations of Patent Law.
- The application is not in conformity with the provisions of Article 20(1) of Patent Law.
- The divisional application is not in conformity with the provisions of Rule 43(1) of the Implementing Regulations of Patent Law.

The detailed analysis for above conclusive opinion is described in the text of this office action.

### 7. On the basis of the above conclusive opinion, the examiner holds that:

- the applicant should make amendment to the application document in accordance with the requirements described in the text of this office action.
- the applicant should, as a response, expound reasons to prove the patentability of this application and amend the application document where it is not in conformity with the related regulations as mentioned in the text of this office action; otherwise the patent right shall not be granted.
- the application does not possess any substantive patentable contents, if the applicant fails to expound reasons or the reasons expounded are not sufficient, this application will be rejected.
- \_\_\_\_\_

### 8. The applicant must pay special attention to the following matters:

- (1) The applicant must submit a response within four months from the date that this office action is received, pursuant to the provisions of Article 37 of Patent Law. If the applicant fails to respond within the time limit without any justified reason, the application will be deemed to have been withdrawn.
- (2) The amendment made by the applicant must not go beyond the scope recorded in the initial description and claims as required by the provisions of Article 33 of Patent Law;

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## THE STATE INTELLECTUAL PROPERTY OFFICE OF THE PEOPLE'S REPUBLIC OF CHINA

meanwhile, the amendment shall be made in accordance with the requirements of this office action as required by the provisions of Rule 51(3) of the Implementing Regulations of Patent Law.

- (3) The applicant's response and/or amendment text must be mailed or submitted to the Receiving Department of the Patent Office of the State IP Office. Documents not mailed or submitted to the Receiving Department do not possess legal effect.
  - (4) The applicant and/or his (its) agent may not come to the Patent Office of the State IP Office to interview with the examiner without an appointment.
9. The text of this office action consists of a total of 5 sheets, and is accompanied by the following annexes:
- the copies of the cited reference documents, total of 1 set and 6 sheets.
- \_\_\_\_\_

Examiner: Fangfang Peng  
Tel: 010-62414402

Examination Department: The Patent Examination Cooperation Center

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(Translated by AFD)



# 中华人民共和国国家知识产权局

**100192**

北京市海淀区学清路8号B座1601A 北京安信方达知识产权代理有限公司  
卢素华 郑霞

发文日:

2010年03月10日



申请号或专利号: 200580002471.5

发文序号: 2010030600050260

申请人或专利权人: 巴特尔纪念研究所

发明创造名称: 生产高铁酸盐(VI)的方法和装置

## 第一次审查意见通知书

(进入国家阶段的PCT申请)

1.  应申请人提出的实质审查请求,根据专利法第35条第1款的规定,国家知识产权局对上述发明专利申请进行实质审查。  
 根据专利法第35条第2款的规定,国家知识产权局决定自行对上述发明专利申请进行实质审查。
2.  申请人要求以其在:  
US专利局的申请日2004年01月16日为优先权日。
3.  经审查,申请人于\_\_\_\_提交的修改文件,不符合专利法实施细则第51条第1款的规定,不予接受。
4.  审查是针对原始提交的国际申请的中文文本或中文译文进行的。  
 审查是针对下列申请文件进行的:  
2006年7月14日提交的摘要附图、说明书摘要、说明书第2,4-11,13-18,20-22,25-31,33-44,47-54,56-68,70-160,162-175,177,180-186,188-232,235-264,266-348段;  
2006年9月5日提交的说明书附图、说明书第144,161,178-179,187,222,233-234段;  
2009年7月17日提交的权利要求书第1-96项、说明书第1,3,12,19,23,24,32,45,46,55,69,176,200,265段。
5.  本通知书引用下列对比文献(其编号在今后的审查过程中继续沿用)

编号	文件号或名称	公开日期 (或抵触申请的申请日)
1	US4256551	19810317

6. 审查的结论性意见:

关于说明书:

- 申请的内容属于专利法第5条规定的不授予专利权的范围。
- 说明书不符合专利法第26条第3款的规定。
- 说明书不符合专利法第33条的规定。
- 说明书的撰写不符合专利法实施细则第17条的规定。

关于权利要求书:



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电子申请,应当通过电子专利申请系统以电子文件形式提交相关文件。除另有规定外,以纸件等其他形式提交的文件视为未提交。



# 中华人民共和国国家知识产权局

- 权利要求\_\_\_\_不符合专利法第2条第2款的规定。  
 权利要求\_\_\_\_不符合专利法第9条第1款的规定。  
 权利要求1-4, 6-7, 12-18不具备专利法第22条第2款规定的新颖性。  
 权利要求4-13, 19-24不具备专利法第22条第3款规定的创造性。  
 权利要求\_\_\_\_不具备专利法第22条第4款规定的实用性。  
 权利要求\_\_\_\_属于专利法第25条规定的不授予专利权的范围。  
 权利要求\_\_\_\_不符合专利法第26条第4款的规定。  
 权利要求1与(46, 62, 93, 96)之间以及25与78之间不符合专利法第31条第1款的规定。  
 权利要求\_\_\_\_不符合专利法第33条的规定。  
 权利要求\_\_\_\_不符合专利法实施细则第19条的规定。  
 权利要求\_\_\_\_不符合专利法实施细则第20条的规定。  
 权利要求\_\_\_\_不符合专利法实施细则第21条的规定。  
 权利要求\_\_\_\_不符合专利法实施细则第22条的规定。

- 申请不符合专利法第26条第5款或者实施细则第26条的规定。  
 申请不符合专利法第20条第1款的规定。

分案申请不符合专利法实施细则第43条第1款的规定。

上述结论性意见的具体分析见本通知书的正文部分。

## 7. 基于上述结论性意见，审查员认为：

- 申请人应当按照通知书正文部分提出的要求，对申请文件进行修改。  
 申请人应当在意见陈述书中论述其专利申请可以被授予专利权的理由，并对通知书正文部分中指出的不符合规定之处进行修改，否则将不能授予专利权。  
 专利申请中没有可以被授予专利权的实质性内容，如果申请人没有陈述理由或者陈述理由不充分，其申请将被驳回。

## 8. 申请人应注意下列事项：

(1) 根据专利法第37条的规定，申请人应当在收到本通知书之日起的4个月内陈述意见，如果申请人无正当理由逾期不答复，其申请将被视为撤回。

(2) 申请人对其申请的修改应当符合专利法第33条的规定，不得超出原说明书和权利要求书记载的范围，同时申请人对专利申请文件进行的修改应当符合专利法实施细则第51条第3款的规定，按照本通知书的要求进行修改。

(3) 申请人的意见陈述书和/或修改文本应当邮寄或递交国家知识产权局专利局受理处，凡未邮寄或递交给受理处的文件不具备法律效力。

(4) 未经预约，申请人和/或代理人不得前来国家知识产权局与审查员举行会晤。

## 9. 本通知书正文部分共有5页，并附有下列附件：

- 引用的对比文件的复印件共1份6页。

审查员：彭芳芳

联系电话：010-62414402

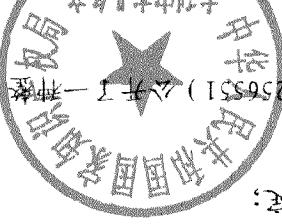
审查部门：专利审查协作中心



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电子申请，应当通过电子专利申请系统以电子文件形式提交相关文件。除另有规定外，以纸件等其他形式提交的文件视为未提交。

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软件申请，回函请寄：100088 北京市海淀区蔚蓝门桥西土城路 6 号 国家知识产权局专利受理处  
电子邮件，应当通过电子邮件系统以电子邮件形式提交相关文件。除另有规定外，以纸件或者其他形式提交的文件视为未提交。



1. 权利要求 1 请求保护一种整体式电化学电解槽，对比文件 1 (US4256531) 公开了一种整体式电化学电解槽，对权利要求 1 具有新颖性的规定。
2. 权利要求 4-13, 19-24 不符合专利法第 22 条第 2 款有关新颖性的规定。
3. 权利要求 1-4, 6-7, 12-18 不符合专利法第 22 条第 2 款有关创造性的规定。

二、依据目前文本，对独立权利要求 1 及其从属权利要求的审查意见如下：

合专利法第 31 条第 1 款的规定为由做出驳回决定。

不符合专利实施细则第 51 条第 3 款规定的，审查员将基于目前审查的申请文本，以本申请不符合专利实施细则第 51 条第 3 款规定的，这种修改是权利要求 1 不具有单一性的未经检索的权利要求 46、62、93、96 及其从属权利要求 1，而保留与该申请人注意的是：对申请文件的修改如果只是删除已检索和评述过的权利要求 1，而保留与该申请已对权利要求 1 及其从属权利要求进行了检索和审查，申请人应当删除与之不具有单一性的技术手段，因而权利要求 1 与 93 之间以及权利要求 1 与 96 之间不具有单一性。

权利要求 1 与权利要求 93、96 的相同或相应技术特征为：一界定了整体式罐的外表，其内装有阴阳极和电解液以及设置电解液进口和出口为所属领域惯用的一整体制备装置的外表，其内装有阴阳极和电解液。但对于电化学电解领域，电解槽具有阴极液体进出口、电解液出口、阴阳极以及电解液。同样的电解槽为本领域很常见的电解槽，为电化学领域惯用的技术手段，因而权利要求 1 与 46 之间以及权利要求 1 与 62 之间不具有单一性。

权利要求 1 与权利要求 46、62 的相同或相应技术特征为：一整体式电化学电解槽（电化学电解槽），其包含一阴极、一阳极以及一种电解质溶液。但包含阴阳极及电解质溶液的整体结构（电解槽），其包含一阴极、一阳极以及一种电解质溶液。因而权利要求 1 与 46 之间以及权利要求 1 与 93-95、VII(96)。具有阴极液体进出口、电解液出口、阴阳极以及电解液。但对于电化学电解领域，电解槽具有阴极液体进出口、电解液出口、阴阳极以及电解液。同样的电解槽为本领域很常见的电解槽，为电化学领域惯用的技术手段，因而权利要求 1 与 46 之间以及权利要求 1 与 62 之间不具有单一性。

一、独立权利要求 1 与权利要求 46、62、93、96 不具有单一性，不符合专利法第 31 条第 1 款的规定。

本申请的权利要求有以下七组：I (1-24)、II (25-45)、III (46-61)、IV (62-77)、V (78-92)、VI (93-95)、VII (96)。

本申请涉及一种电化学电解槽及其操作方法，以及用于制造高纯度盐的方法，经审查，现提出如下的审查意见。

申请号：200580024715

(进入国家阶段的 PCT 申请)

## 第一次审查意见通知书

中华人民共和国国家知识产权局





# 中华人民共和国国家知识产权局

体式电化学电解槽，其内部无隔膜，具有界定一整体式室的外壳，外壳具有一电解液进口7，伴随盐水的液氯出口8、伴随盐水的氢氧化钠出口9和氢气出口10，腔室内设置有阴极17和阳极18以及内部装载有电解液，所述的阴极可以为镍或低碳钢网，阳极可以为钛网（阴阳极均是非气体扩散性电极）（参见对比文件1的摘要，说明书第1-2栏以及第4栏第39行至第5栏第42行，附图1, 2, 3），由此可见，对比文件1已经公开了该权利要求的全部技术特征，两者的技术方案相同，且对比文件1所公开的技术方案与该权利要求请求保护的技术方案属于相同的技术领域，并且采用相同的技术方案，能够解决同样的技术问题，并能够产生相同的技术效果，因此该权利要求请求保护的技术方案不具备新颖性。

2、权利要求2为权利要求1的从属权利要求，其附加的技术特征是对电解液出口作了进一步的限定，但该附加技术特征已被对比文件1公开了（参见对比文件1的说明书第5栏第22-42行，附图1, 2, 3）：电解槽外壳设有伴随盐水的液氯出口8、伴随盐水的氢氧化钠出口9，即具有两个电解液出口。因此，在其引用的权利要求不具备新颖性的基础上，该权利要求也不具备新颖性。

3、权利要求3、4分别为权利要求1、3的从属权利要求，但其附加的技术特征已被对比文件1公开了（参见对比文件1的说明书第5栏第1-42行，附图1, 2, 3）：电解槽中设置有分流器19（作为一种堰板结构的流体流量控制器），与电解液出口流体相通，控制电解液流出出口的量以控制盐水电解液在阴极和阳极表面的流量。因此，在其引用的权利要求不具备新颖性的基础上，权利要求3、4也不具备新颖性。同时，流体流量控制器选自限流器、阀、格栅、流体流量收缩管道、曲管也仅是本领域的常规选择，即当选用上述流体流量控制器时，权利要求4不具备创造性。

4、权利要求5为权利要求1的从属权利要求，其附加的技术特征是对电解槽中阳极材料作了进一步的限定。但对于本领域技术人员来讲，选用含铁的材料制成的阳极为本领域的常规选择，并不需要付出创造性的劳动，因此，在其引用的权利要求不具备新颖性的基础上，该权利要求不具备创造性。

5、权利要求6、7为权利要求1的从属权利要求，其附加的技术特征是对电解槽中阳极作了进一步的限定。但该附加技术特征已被对比文件1公开（参见对比文件1的说明书第2栏第18-31行及第5栏第1-9行）：电解槽中阳极可以为钛网（多孔金属网的下位概念），<sup>例如可以是包裹氯化钛的多孔金属网</sup>

2010.2 电子申请：应当通过电子邮件申请系统以电子邮件形式提交相关文件。除另有规定外，以纸件或者其他形式提交的文件视为未提交。

9、权利要求19为权利要求1的从属权利要求，权利要求20、21分别为权利要求19的从属权利要求，权利要求22为权利要求20的从属权利要求。对比文件1公开的电解槽中为变化阳极表面的电解液流量可设置有分流器，例如可以控制阳极表面流量为 $40\text{mL}/\text{min}$ ，阳极表面流量为 $30\text{mL}/\text{min}$ (参见对比文件1的第5栏第1-42行以及实施例)，为了更好的控制阳极表面不同的电解液流量，在电解槽的阴阳极间设置带有孔格的格栅对于本领域技术人员来说是显而易见的，且为抗碱液的腐蚀，选用聚烯烃、含氟聚合物或聚氯乙烯的塑料作为格栅材料不仅为防腐蚀且为常规选择，并不需要付出创造性的努力，同时格栅上筛孔提供阴阳极之间的流通开敞区。

8. 权利要求14为权利要求14的从属权利要求，权利要求15、16、17为权利要求14的从属权利要求，权利要求18为权利要求17的从属权利要求，其附加的技术特征仅是对方解码中所载的工作波作了进一步的限定，但对比文件1中所述对方解码电路载有该工作波，附加技术特征中对工作波作了进一步的限定，并不能将请求保护的对方解码结构与对比文件1中的对方解码区分开，因此，在其引用的对方解码的限定并不具备新颖性。权利要求14-18也不具备新颖性。

7. 权利要求 12、13 从属权利要求 1 的从属权利要求，其附加的技术特征是对电解槽中阴极进行了进一步的限定。但对比文件 1 已公开了该附加技术特征（参见对比文件 1 的第 5 段第 1-9 行）：电解槽的阴极可以是镍或低碳钢网。因此，在其引用的权利要求不具备新颖性的基础上，行）：电解槽的阴极可以是镍或低碳钢网。因此，在其引用的权利要求不具备新颖性的基础上，权利要求 12、13 也不具备新颖性。同时，阴极选用阳极材料特征中的其他材料或结构时，权利要求 12、13 也并不具备创造性，它通过本领域的常规选择即能获得。

6、权利要求8、9均为权利要求1的从属权利要求，权利要求10、11均为权利要求9的从属权利要求，其附加的技术特征是对阴极的相对表面面积作了进一步的限定。但对于本领域的技术人员来讲，经过很常规的实验调整就能够获得上述附加技术特征中相应的阴极表面积比而言并不需要付出创造性的劳动。因此，在其引用的权利要求不具备新颖性/创造性的基础上，权利要求8、9、10、11不具备创造性。

化铁、氯化钙的铁网（一种DSA电极）。因此，在其引出用的权利要求不具新颖性的基础上，权利要求6、7也不具备新颖性。另外，阳极选用固体铁粉、钢丝网、编制金属网布、金属丝、镍或其组合，铁、钢、铂、钛或其他抗氧化化的导电材料对于本领域技术人员来讲，也是很容易就能想到的，即这种情况下，权利要求6、7不具备创造性。





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的大小经过本领域技术人员的有限次实验调整即能获得。因此，在其引用的权利要求不具备新颖性/创造性的基础上，权利要求19-22不具备创造性。

10、权利要求23为权利要求1的从属权利要求，其附加的技术特征是对电解槽外壳作了进一步的限定。对比文件1公开的作为电解槽外壳的底、壁及盖可选用玻璃或硅材料（参见对比文件1的说明书第4栏第51-68行），在该公开内容的基础上，将外壳由选自金属、玻璃纤维、热塑性增强塑料或者热固性增强塑料、混凝土、橡胶或者其组合、包括构造成刚性壁结构内的抗腐蚀内衬的材料来制备对于本领域技术人员来讲是很容易想到的。因此，在其引用的权利要求不具备新颖性的基础上，权利要求23不具备创造性。

11、权利要求24为权利要求1的从属权利要求，其附加的技术特征是对电源作了进一步的限定，但选用可变直流电源，可操作地连接到电化学电解槽上为所属领域技术人员的常用技术手段，并不需要付出创造性的劳动。因此，在其引用的权利要求不具备新颖性的基础上，权利要求24不具备创造性。

### 三、独立权利要求25与权利要求78之间缺乏单一性，不符合专利法第31条第1款的规定。

权利要求25与权利要求78相同或相应的特征为：电化学电解槽具有一界定一整体式室的外壳，外壳具有一个电解液进口、至少两个电解液出口，室内有阴阳极、从进口引入的电解质溶液、控制电解液流出电解液出口的量以使流过阳极的电解液比流过阴极的多。由前述审查意见1、3、9可以看出上述技术特征已被对比文件1公开（在此不再重复），即并不是对现有技术作出贡献的技术特征，因此权利要求25、78之间不存在共同的或相应的特定技术特征，从而缺乏单一性。

申请人应当在本通知书指定的答复期限内对本通知书提出的问题逐一进行答复，必要时应修改专利申请文件，否则本申请将难以获得批准。申请人对申请文件的修改应当符合专利法第三十三条的规定，不得超出原说明书和权利要求书记载的范围。

申请人提交的修改文件应当包括：修改涉及部分的原文复印件，在该复印件上标注出所作的增加、删除或替换；重新打印的替换页（一式两份），用于替换相应的原文。申请人应当确保上述两部分在内容上的一致性。

210402 纸件申请，回函请寄：100088 北京市海淀区蓟门桥西土城路6号 国家知识产权局专利局受理处收  
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电子邮件，应当通过电子专利申请系统以电子文件形式提交相关文件。除另有规定外，以纸件等其他形式提交的

审查员代码:199121

审查员姓名:姚芳芳



# United States Patent [19]

Cliff et al.

[11]

4,256,551

[45]

Mar. 17, 1981

[54] ELECTROLYTIC PROCESS

[75] Inventors: Michael J. Cliff, Elton near Chester; William T. Cross, Cuddington; Colin Ramshaw, Norley, all of England

[73] Assignee: Imperial Chemical Industries Limited, London, England

[21] Appl. No.: 87,569

[22] Filed: Oct. 23, 1979

[30] Foreign Application Priority Data

Nov. 2, 1978 [GB] United Kingdom 42888/78

[51] Int. Cl. 3 C25B 1/14; C25B 1/26;  
C25B 11/03

[52] U.S. Cl. 204/98; 204/278

[58] Field of Search 204/98, 266, 278

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[57] ABSTRACT

A process for electrolyzing at least a proportion of a liquid comprising an electrolyte which process comprises passing the liquid between electrodes, at least one of which is an anode and at least one of which is a cathode, the liquid being in a state of laminar flow substantially parallel to the electrodes, the electrodes being disposed substantially horizontally above each other and each electrode being permeable to the product(s) or solutions thereof produced at or adjacent the electrode and, where a density difference is generated on production of the products, the relative disposition of the electrodes is such that the density difference reduces the possibility of the product(s) produced at or adjacent the anode mixing with the product(s) produced at or adjacent the cathode. The process is especially applicable to the electrolysis of brine.

15 Claims, 4 Drawing Figures

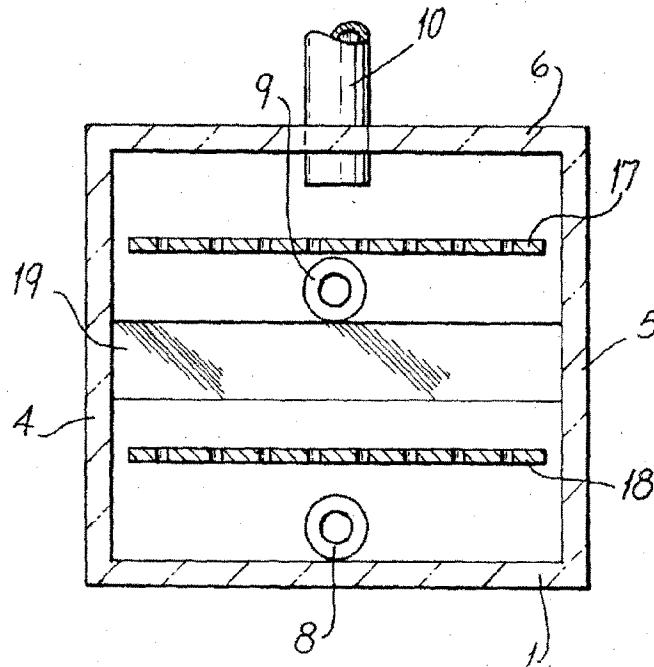


Fig. 1.

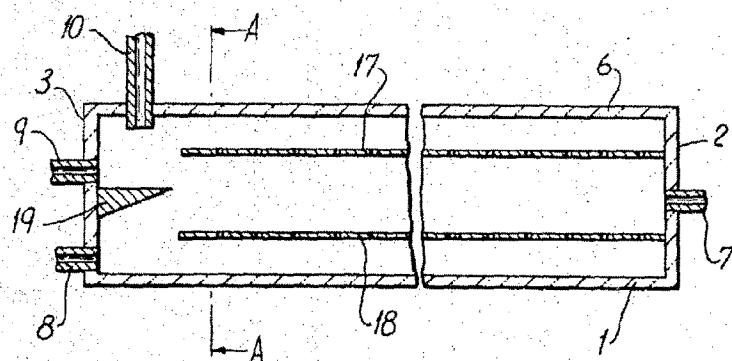


Fig. 2.

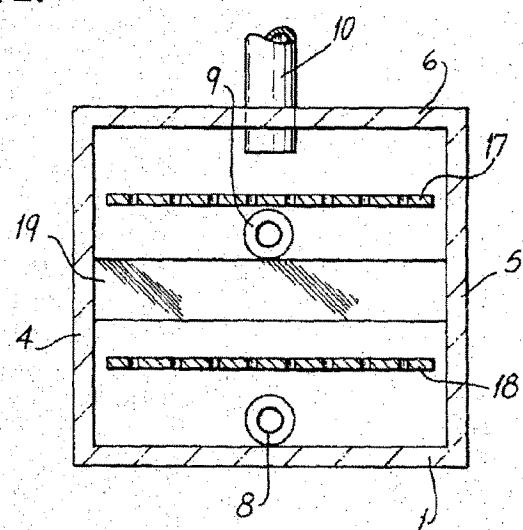


Fig. 3.

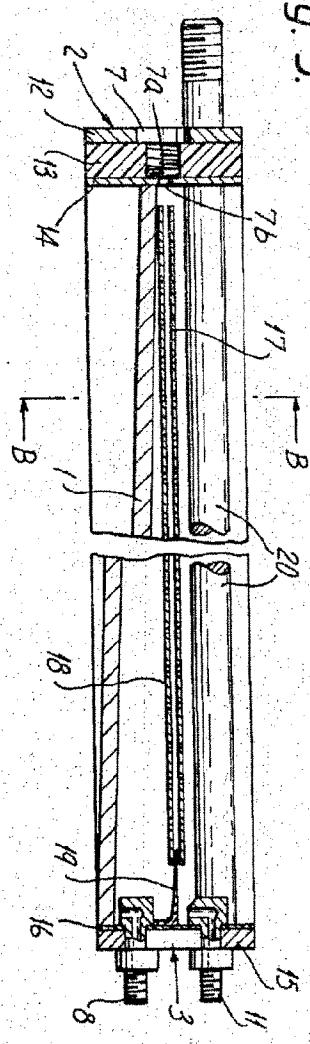
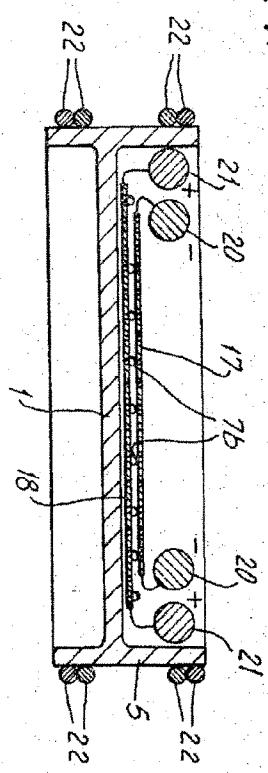


Fig. 4.



## ELECTROLYTIC PROCESS

This invention relates to electrochemical processes, particularly to electrolytic processes and to apparatus in which such processes are carried out.

In the electrolysis of an electrolyte in a liquid in an electrolytic cell, often a porous partition, typically a permeable membrane, is disposed between the anode and the cathode to prevent or reduce the product(s) produced at the anode mixing or reacting with the products(s) produced at the cathode. Moreover where an electrolytic product is immiscible with the liquid it often forms a polarising layer on the electrode at which it is produced, which layer hinders or interrupts the electrolysis. We have now found that where the liquid flows under conditions of laminar flow substantially parallel to the surface of substantially horizontal electrodes, these problems can be alleviated by employing electrodes which are permeable to the product(s) or solution(s) thereof produced at or adjacent the electrodes.

Accordingly, the present invention provides a process for electrolysing at least a proportion of a liquid comprising an electrolyte which process comprises passing the liquid between electrodes at least one of which is an anode and at least one of which is a cathode, the liquid being in a state of laminar flow substantially parallel to the electrodes, the electrodes being disposed substantially horizontally above each other and each electrode being permeable to the product(s) or solutions thereof produced at or adjacent the electrode and, where a density difference is generated on production of the products, the relative disposition of the electrodes is such that the density difference reduces the possibility of the product(s) produced at or adjacent the anode mixing with the product(s) produced at or adjacent the cathode.

While the liquid may be a molten electrolyte preferably it is a solution or dispersion of the electrolyte in a suitable solvent.

By "electrolyte" we mean a substance which in a suitable solvent or gel or as a melt gives rise to at least one anion and at least one cation. An electrolyte may be a so-called "strong" electrolyte, i.e. an electrolyte which contains a stable ionic bond and is substantially wholly ionised in solution. An electrolyte may be a so-called "weak" electrolyte, i.e. an electrolyte which contains a covalent bond which may be transformed into an ionic bond such that a solution of the electrolyte in a suitable solvent or a melt of the electrolyte contains ionic and covalent bonds in dynamic equilibrium. Electrolytes are typically acids, bases or salts.

By "electrolysis" we mean the decomposition of an electrolyte by the passage of an electric current through a solution or a melt of the electrolyte such that at least one anion migrates to an anode to lose at least a proportion of its charge and at least one cation migrates to a cathode to lose at least a proportion of its charge.

Throughout this specification, where reference is made to "electrode", "anode" or "cathode" such expressions are intended to include the case where a plurality of anodes and/or cathodes are present.

By laminar flow we mean flow of a liquid in parallel layers in contact with each other with little or no fluctuation or turbulence disturbing the layers.

By "product produced at or adjacent an electrode" we mean (a) the uncharged atom or group which may

be deposited on or liberated at the electrode to which an ion migrates and gives up its electric charge, e.g. an anion migrates to an anode and loses an electron to the anode, and a cation migrates to a cathode and accepts an electron from the cathode, or (b) a product formed when the uncharged atom or group reacts chemically with the electrode, a solvent (where it is present), or with a substance present in the liquid.

The permeable electrodes may be formed of parallel, woven, knitted or sintered fibres, sintered particles, perforated plate, expanded metal or a skeleton foam, or mechanical assemblies thereof. Preferably the electrode is a woven mesh or a plurality of parallel fibres or an expanded metal. Preferably the "pores" of such electrodes or, where parallel fibres are employed the distance between fibres, are smaller than the distance between the anode and the cathode.

The material of the electrodes will necessarily be electrically conducting and will be chosen with regard to the liquid and product(s) which they are to contact and the mechanical stresses to which they are to be subjected. Suitable materials are well known in the electrolytic art and choice of suitable materials will present no problem to the skilled man. As examples we would mention a nickel or mild steel cathode and a titanium anode for the electrolysis of an alkali metal halide brine, e.g. sodium chloride brine. It will be appreciated that to facilitate disengagement of a product from an electrode the electrode may have a suitable surface treatment which inhibits the product wetting the electrode. It will be appreciated that to facilitate discharge of ions at an electrode it may have a suitable surface treatment. However, such treatments should not catalyse or facilitate an undesirable reaction. For example, in the electrolysis of brine the anode may be treated with titanium/ruthenium oxides which facilitate the discharge of chloride ions but does not give a sufficiently high over-voltage for the production of oxygen.

For a given liquid at a given temperature and a given rate of flow, the upper limit of the distance between the anode and the cathode (the anode/cathode gap) is dictated by the Reynolds Number at which turbulence sets in and by the acceptable ohmic drop between the anode and the cathode. Likewise as the anode/cathode gap is decreased the practical difficulty of maintaining them parallel increases and imposes a lower limit on the said gap. Preferably the anode/cathode gap is between 0.1 mm and 2 cm, more preferably between 0.5 mm and 4 mm.

As electrolysis proceeds the density of the liquid in the region of the electrodes may change and the change of density may be different in the region of the anode to that in the region of the cathode. The change in the density of the liquid in the region of an electrode is dependent inter alia on the loss of electrolyte from the said region and on the nature of the product produced in the said region, e.g. its miscibility or lack of miscibility with the liquid.

The voltage drop between the anode and the cathode due to the ohmic resistance of the solution, where a solution is employed, will depend inter alia, on the particular electrolyte present and on the ionic concentration thereof. Furthermore, it will be appreciated that in order to reduce energy consumption the voltage drop is kept as low as possible commensurate with an acceptable rate of production of products. The voltage drop may be reduced by increasing the ionic concentration of the electrolyte or by increasing the temperature. This

may necessitate operating at the saturation concentration of an electrolyte in a solvent. For example, the electrolysis of sodium chloride is typically effected in saturated brine.

A product of the process of the present invention may be a gas, a liquid or a solid. The process may be operated under conditions of temperature and pressure such that the product, where it is a gas at ambient temperature and pressure, is produced as a liquid. For example, where chlorine is produced at the anode by the electrolysis of an alkali metal chloride brine the temperature of the brine is kept in the range 10° C.-100° C., preferably in the range 30° -50° C., and the pressure is kept in the range 50 to 750 psi, preferably in the range 130-500 psi such that the chlorine is obtained as a liquid.

Where a change in density occurs in the region of an electrode the relative disposition of the electrodes is chosen such that the change in density reduces the possibility of the product(s) produced at or adjacent the said electrode mixing with the product produced at or adjacent the electrode of opposite polarity. For example, where the density of a product or solution thereof produced at or adjacent a cathode has a density lower than that of the liquid, the cathode is disposed above the anode, e.g. where brine flows horizontally between an anode and a cathode, the cathode is made the upper electrode such that hydrogen produced may be readily disengaged from the liquid. Moreover, where the electrolysis of brine is effected under conditions of temperature and pressure such that liquid chlorine is produced, which has a higher density than that of brine or caustic soda, disposition of the anode below the cathode further reduces the possibility of the products mixing.

When liquid product e.g. liquid chlorine is produced at the lower electrode it is preferred that the depth of liquor below the lower electrode should increase through the cell in the direction of liquid electrolyte flow. Conveniently, this may be achieved by sloping the base of the cell downwardly towards the outlet for the liquid product.

Where a gas is produced at the upper electrode the depth of the liquid above the upper electrode is preferably kept to a minimum to allow ready disengagement of the gas. For example in the electrolysis of brine the depth of liquid above the upper electrode (the cathode) is preferably between 0.2 mm and 3 mm, more preferably between 0.5 mm and 1.5 mm, to allow ready disengagement of the hydrogen produced at the cathode. We have found that where a gas is produced at the upper electrode the height of liquid above the upper electrode may conveniently be kept at a pre-set height by positioning the gas take-off port at the pre-set height with liquid discharging through the gas take-off port such that if the rate of flow of liquid increases the pressure of the gas produced forces excess liquid out of the gas take-off port.

It will be appreciated that the density of the layers of liquid adjacent the electrodes may be altered by imposing a temperature gradient on the cell in which the process of the present invention is carried out which temperature gradient may reduce the possibility of a product produced at or adjacent the anode mixing with the product produced at or adjacent the cathode. For example, where brine is electrolysed as it flows horizontally between the electrodes the upper of which is the cathode, the region of the cathode is preferably heated to lower the density of the caustic soda produced in the said region so that the possibility of the caustic soda

drifting to the anode is reduced. The distance over which the liquid flows in contact with the electrodes in the process of the present invention is dependent inter alia on the rate of flow of liquid, and on the current density, i.e. the faster the rate of flow the longer the cell and the higher the current density the shorter the cell. At convenient rates of flow and current density the length of the cell is typically between 5 and 50 cm, preferably between 15 and 30 cm. The current density employed will depend on the reaction occurring in the cell and for the electrolysis of brine we have found that current densities between 0.1 and 2.0 amp/cm<sup>2</sup> and preferably between 0.2 and 0.6 amp/cm<sup>2</sup> may be employed.

The rate of flow of the liquid is chosen such that it is sufficient to produce product(s) at an acceptable rate, to maintain a suitable temperature in the cell, and to maintain laminar flow.

The electrolytic process according to the present invention may be employed inter alia in the production of chemicals, e.g. hydrogen, oxygen, hydrogen peroxide, chlorine caustic soda, fluorine; in the extraction of metals from molten salts, e.g. aluminium, magnesium, sodium and from solutions of metal ores, e.g. copper, zinc, cadmium.

The invention will be further described by reference to the accompanying drawings which show, by way of example only, two chlorine cells suitable for use in the process of the present invention. In the drawings:

FIG. 1 is a vertical longitudinal section through a first cell;

FIG. 2 is a cross-section on the line AA of FIG. 1 to a different scale;

FIG. 3 is a vertical longitudinal section through a second cell having a modified exit port and showing further details of construction.

FIG. 4 is a cross-section on the line BB of FIG. 3.

Referring to FIGS. 1-4, each cell is provided with a baseplate 1, end walls 2,3 (shown diagrammatically in FIG. 2 and in more detail in FIG. 4), sidewalls 4,5, and a separate cover 6 (as shown in FIGS. 1 and 2) but which may alternatively be effected by superimposing two cells (of the type shown in FIGS. 3 and 4) on top of one another. The baseplate 1, end walls 2, 3, sidewalls 4, 5 and cover 6 are suitably fabricated of glass or silica. The baseplate 1 slopes downwardly (as shown in FIG. 3; not shown in FIG. 1) from side wall 2 to end wall 3 at a shallow angle, for example at an angle to the horizontal of from 1° to 10° typically 2°.

Each cell is provided with an inlet port 7 for sodium chloride brine, and an outlet port 8 for liquid chlorine. The inlet port 7 (as shown in FIG. 3) is conveniently connected with a header 7a from which it is fed to the cell through a plurality of ports 7b. The cell shown in FIGS. 1 and 2 has an outlet port 9 for sodium hydroxide solution and an outlet port 10 for hydrogen. The cell shown in FIGS. 3 and 4 has a single outlet port 11 for both sodium hydroxide solution and hydrogen. The inlet port 7 and the outlet ports 9, 10, 11 are typically of mild steel, and the outlet port 8 is typically of titanium.

The end wall 2 (as generally indicated in FIG. 3) typically comprises a mild steel end plate 12, a block or slab 13 of plastics material (e.g. polyvinyl chloride, polytetrafluoroethylene) and a thin sheet 14 of titanium provided with ports 7b connecting with header 7a and inlet port 7 (as generally indicated in FIG. 3). The end wall 3 (shown in FIG. 3) typically comprises a mild

steel end plate 15, and a sheet 16 of plastics material (e.g. polyvinyl chloride, polytetrafluoroethylene).

A cathode 17 is typically formed of nickel or mild steel mesh. Anode 18 is typically formed of titanium mesh and is provided with an electrocatalytically active coating, for example a coating comprising a mixture of ruthenium oxide and titanium dioxide. A splitter 19, typically of titanium serves to vary the flow of brine over the anode and cathode surfaces respectively.

Current is fed to the cathode 17 by means of copper busbars 20 and to the anode 18 by means of copper busbars 21 (as shown in FIGS. 3 and 4; the electrical leads are not shown in FIGS. 1 and 2). The busbars 20, 21 may be connected to the cathode 17 and anode 18 by any convenient means, for example by brazing or clamping, and may be protected from conditions within the cell environment by suitably plating, for example with nickel.

The end plates 2, 3 are conveniently held together by means of tie rods 22 (as shown in FIG. 4) typically of mild steel.

In the electrolysis of brine in the cell, brine (6N) at a temperature of 30° C. flows in through port 7 to develop a pressure of 150 to 250 psi, e.g. 200 psi, in the cell and flows through the cell under conditions of laminar flow. Chlorine produced at the anode 18 is formed as a liquid and falls to the bottom of the cell and is expelled from the cell via port 8 along with brine. Sodium hydroxide solution is produced at cathode 17 and is discharged with brine through port 9 (FIG. 1) or port 11 (FIG. 3). Hydrogen is produced at the cathode; it collects at the top of the cell and escapes through port 10 (FIG. 1) or port 11 (FIG. 3). The pressure of hydrogen can be used to keep the level of liquid in the cell below a pre-set height indicated by the dotted line in FIGS. 1 and 3, e.g. if the flow of brine increases so that the level tends to rise above the dotted line, the pressure of the hydrogen forces excess brine out through the port 10 (FIG. 1) and port 11 (FIG. 3).

The invention is further illustrated by the following Example relating to the electrolysis of sodium chloride brine.

#### EXAMPLE

The cell (20 cm long and of cross-section 2 cm by 2 cm) was provided with a titanium mesh anode coated with a mixture of ruthenium oxide and titanium dioxide and a titanium mesh cathode which was similarly coated (the coating served to protect the titanium cathode from hydrogen attack).

6N sodium chloride brine was passed between the electrodes at a rate of 70 ml/min (split approximately 40 ml/min to the anode surface and 30 ml/min to the cathode surface). The cell was maintained at a pressure of 205 psi, and operated at 3.4 volts initially (which gradually increased to 3.7 volts over 3 hours) and at a current density of 0.25 amp/cm<sup>2</sup>. The anode/cathode gap was 4 mm and the depth of brine above the cathode was about 1.5 mm. Liquid chlorine and dissolved chlorine, totalling 0.088 M in brine, was discharged from the bottom of the cell. Sodium hydroxide (0.01 M in brine), hydrogen and chlorine (0.01 M in brine) were discharged from the top of the cell.

What we claim is:

1. A process for electrolysing at least a proportion of 65 a liquid comprising an electrolyte which process comprises passing the liquid between electrodes, at least one of which is an anode and at least one of which is a

cathode, the liquid being in a state of laminar flow substantially parallel to the electrodes, the electrodes being disposed substantially horizontally above each other and each electrode being permeable to the product(s) or solutions thereof produced at or adjacent the electrode and, where a density difference is generated on production of the products, the relative disposition of the electrodes is such that the density difference in combination with laminar flow of the liquid reduces the possibility of the product(s) produced at or adjacent the anode mixing with the product(s) produced at or adjacent the cathode.

2. A process as claimed in claim 1 wherein the anode/cathode gap is between 0.1 mm and 2.0 cm.

3. A process as claimed in claim 2 wherein the anode/cathode gap is between 0.5 mm and 4 mm.

4. A process as claimed in any one of the preceding claims wherein each electrode comprises a woven mesh or a plurality of parallel fibres or an expanded metal.

5. A process as claimed in claim 4 wherein the pores of the electrode or, where parallel fibres are employed, the distance between fibres, are smaller than the anode/cathode gap.

6. A process as claimed in any one of the preceding claims wherein the electrodes are disposed so that the product(s) or solutions thereof produced at or adjacent to the lower electrode having a density which is greater than that of the liquid or other liquid product(s) and so that the product(s) or solutions thereof produced at or adjacent to the upper electrode have a density which is lower than that of the liquid or other liquid product(s).

7. A process as claimed in any one of the preceding claims wherein a temperature gradient is imposed between the electrodes to reduce the possibility of the product(s) or solutions thereof produced at or adjacent to the anode mixing with the product(s) or solutions thereof produced at or adjacent to the cathode.

8. A process as claimed in any one of the preceding claims wherein the pressure and temperature are such that at least one of the products produced at the lower electrode, which is a gas at ambient temperature and pressure, is produced as a liquid having a density greater than that of the liquid or other liquid product(s).

9. A process as claimed in claim 8 wherein the electrolysis is carried out at a temperature in the range 10° C. to 100° C. and at a pressure within the range 50 psi to 750 psi.

10. A process as claimed in claim 9 wherein the electrolysis is carried out at a temperature in the range 30° C. to 50° C. and at a pressure within the range 130 psi to 500 psi.

11. A process as claimed in any one of claims 8 to 10 wherein the depth of liquor below the lower electrode increases through the cell in the direction of liquid electrolyte flow.

12. A process as claimed in any one of the preceding claims wherein at least one of the products produced at the upper electrode is a gas and wherein the depth of liquid above the upper electrode is kept to a minimum to allow ready disengagement of gas.

13. A process as claimed in claim 12 wherein the depth of liquid above the upper electrode is between 0.2 mm and 3 mm.

14. A process as claimed in any one of the preceding claims wherein the liquid is an alkali metal halide brine.

15. A process as claimed in claim 14 wherein the alkali metal halide is sodium chloride.

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